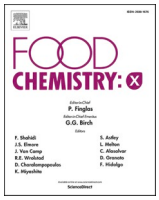
Food Chemistry: X 17 (2023) 100554

Contents lists available at ScienceDirect

Food Chemistry: X

journal homepage: www.sciencedirect.com/journal/food-chemistry-x



Synergy of physicochemical reactions occurred during aging for harmonizing and improving flavor

Wei Jia [a,b,*], Rutian Ma [a], Liangbin Hu [a], Haizhen Mo [a,b,*]

[a] School of Food and Biological Engineering, Shaanxi University of Science & Technology, Xi'an 710021, China
[b] shaanxi Research Institute of Agricultural Products Processing Technology, Xi'an 710021, China

ARTICLE INFO

Keywords:
Baijiu
Aging
Flavor
Mechanism
Aging discrimination

ABSTRACT

Numerous counterfeit vintage Baijiu are widely distributed in the market driven by economic interest which disturb the market economic rules and damage the reputation of particular Baijiu brand. Found on the situation, the Baijiu system variation during aging period, aging mechanisms and discrimination strategies for vintage Baijiu are systematically illuminated. The aging mechanisms of Baijiu cover volatilization, oxidation, association, esterification, hydrolysis, formation of colloid molecules and catalysis by metal elements or other raw materials dissolved from storage vessels. The discrimination of aged Baijiu has been performed by electrochemical method, colorimetric sensor array or component characterization coupled with multivariate analysis. Nevertheless, the characterization of non-volatile compounds in aged Baijiu is deficient. Further research on the aging principles, more easy-operation and low-cost discrimination strategies for aged Baijiu are imperative. The above information is favorable to better understand the aging process and mechanisms of Baijiu, and promote the development of artificial aging techniques.

1. Introduction

Chinese Baijiu, with a time-honored history more than two thousand years, is one of the six world-renowned distilled liquors (Jia, Fan, Du, Shi, & Ren, 2022b). Baijiu has become an essential alcoholic beverage in daily life and occupies significant economic status in domestic food market (Fig. 1) (Wang et al., 2022a). The production process of Baijiu generally covers the selection of raw materials, preparation of starter culture (*Daqu*, *Xiaoqu* or *Fuqu*), saccharification and fermentation, solid-state distillation, aging and blending (Fig. 2a). The freshly distilled Baijiu possesses undesirable flavor characteristics which can be described as peppery, harsh, pungent, green, and unpleasant (He et al., 2021). It is indispensable to store freshly distilled Baijiu in a container for several years to eliminate undesirable odors and make its flavor more harmonious and balanced. The above operation is named aging.

The aging process of Baijiu involves several physicochemical reactions which include volatilization of low boiling point components, association of water and ethanol molecules, redox reaction, esterification and hydrolysis to enhance the quality and acceptability of Baijiu (Dong et al., 2019). Nevertheless, the present proposed aging mechanisms are unconvincing due to the lack of adequate supporting information. The organoleptic quality of Baijiu has a direct impact on consumers' selection and acceptability (Sun, Wang, & Sun, 2021). For the last several decades, various brands and grades of Baijiu are widely distributed in the market (Li et al., 2020a). In general, the common counterfeit method is the substitution of Baijiu with shorter aging years for another aged properly (Li et al., 2021a). Considering the situation, the discrimination of Baijiu with various aging years is crucial for quality control and brand prestige protection of Baijiu.

The traditional aging technology of Baijiu, storing fresh liquor in pottery jars or stainless-steel vessels, has the deficiency of time-consuming, labor-intensive, high storage losses and is not beneficial to the modernization and technological advancement of Baijiu industry (Zhang et al., 2021b). Hence, more comprehensive understanding of natural aging mechanisms of Baijiu and variation of significant characteristic compounds during aging period could aid to the development of innovative artificial aging technologies which are conducive to enhancing process efficiency and reducing cost. From the above perspectives, the available knowledge on the composition variation during Baijiu aging including non-volatile compounds, the aging mechanisms associated with different aging methods, and novel discrimination strategies for Baijiu with various vintage were systematically clarified in

* Corresponding authors at: School of Food and Biological Engineering, Shaanxi University of Science & Technology, Xi'an 710021, China.
E-mail address: jiawei@sust.edu.cn (H. Mo).

https://doi.org/10.1016/j.fochx.2022.100554
Received 23 September 2022; Received in revised form 20 December 2022; Accepted 23 December 2022
Available online 24 December 2022

this article from different aspects.

2. Baijiu system variation during aging period

The variation of Baijiu occurred during aging process is complex and has a prominent influence on the flavor of Baijiu. Aging procedure could eliminate unpleasant flavor compounds such as sulfide, acetaldehyde, acrolein, alkenes, methanol, free ammonia and accompanied with the formation of aromatic compounds through hydrolysis, oxidation, association and esterification (Tan, Zhong, Zhao, Du, & Xu, 2019; Tian et al., 2022). The flavor of Baijiu is the result of synergy reaction of all compounds (Liu & Sun, 2018). Except for volatile compounds which were discussed mostly, non-volatile components also hold an important place in the flavor of Baijiu (Hong, Zhao, & Sun, 2021; Wang, Chen, Wu, & Zhao, 2022b). Currently, the research on the variation of flavor substances in the aging process of Baijiu mainly focused on *Light*-, *Strong*- and *Feng*-flavor Baijiu.

The differences of aroma compounds between fresh and aged *Xiaoqu* Baijiu (*Light*-flavor) were performed through sensomics approach which include gas chromatography-mass spectrometry (GC–MS), aroma extraction dilution analysis, aroma recombination and omission experiments. It could be concluded that further quantity and intensity of aroma-active compounds existed in vintage than fresh *Xiaoqu* Baijiu. The odor activity value of ethyl isovalerate, ethyl butanoate and ethyl

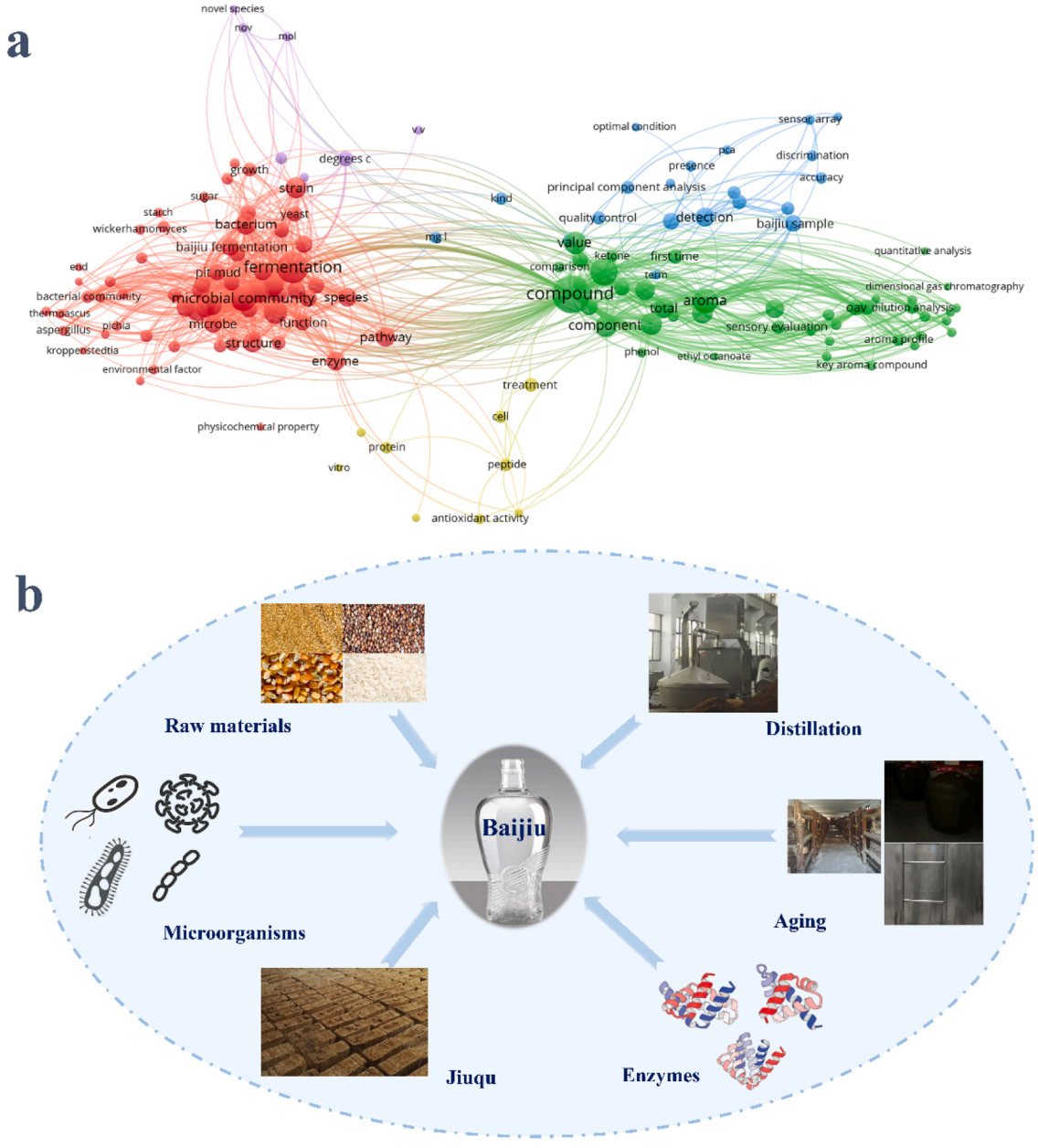


Fig. 1. The current research status of Baijiu and factors affecting the flavor and quality of Baijiu. (a) Correlation network among the current research status of Baijiu: The research mainly focused on multiple microbial community and fermentation process (red circle nodes), flavor profile (green circle nodes) and quality control (blue circle nodes) of Baijiu. (b) Factors affecting the flavor and quality of Baijiu. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

acetate in fresh distilled Baijiu were much lower than aged. Aroma omission experiments suggested that 3-hydroxy-4,5-dimethyl-2(5H)-furanone (sotolon), vanillin and 3-(methylthio)-propionaldehyde (methional) played critical parts in the overall aroma characteristics of aged *Xiaoqu* Baijiu, while sotolon and methional were only distinguished in aged *Xiaoqu* Baijiu (Sun et al., 2022). 1,1-diethoxyethane, 3-methylbutanal, dimethyl trisulfide, ethyl acetate, and ethyl isovalerate were also recognized as significant aroma substances of aged *Xiaoqu* Baijiu.

The flavor-active components of six *Niulanshan* Baijiu (*Light*-flavor) samples with different storage years (3, 4, 5, 6, 8 and 13 years) were systematically analyzed by using a sensomics strategy. A total of 59 odorants were identified, ethyl 2-hydroxy-4-methylvalerate, 1,2-dimethoxybenzene, 1,1-diethoxy-3-methylbutane, ethyl 3-methylthiopropionate and ethyl acrylate were detected for the first time in *Light*-flavor Baijiu. The recombination and omission experiments concluded that ethyl acetate, ethyl acrylate, ethyl 2-methylbutyrate, γ-nonalactone, ethyl isovalerate, ethyl butyrate, isoamyl acetate, ethyl caprylate, ethyl valerate, 3-methylbutanal, β-damascenone, and geosmin have great contribution to the flavor of *Niulanshan* Baijiu (Wang et al., 2022d). The majority of key aroma odorants reached the highest concentration at the longest aging time explored in this study (13 years), while the concentration was relatively stable in the fourth and fifth aging years. The volatile fractions of Chinese *Highland Qingke* Baijiu (*Light*-flavor) with different aging months were observed and analyzed (Wang et al., 2022c). Twelve marker compounds, including seven acetals, two esters, two ketones and 1,3,5-trioxane, were accurately identified through numerous statistics analysis methods. Among them, 1,1-diethoxy-propane, 1,1-diethoxy-butane and 1,1-diethoxy-3-methyl-butane are regarded as significant contributors to the flavor of Baijiu.

The concentration of flavor compounds in *Luzhoulaojiao* Baijiu (*Strong*-flavor) during three years' storage in pottery jars were inquired by means of various chromatography techniques. Eighty-six flavor compounds were determined while esters had the topmost concentration followed by acids, alcohols, aldehydes and ketones. Surprisingly, the total concentration of flavor components slowly reduced during storage. The four backbone components of *Strong*-flavor Baijiu, ethyl acetate, ethyl butanoate, ethyl hexanoate and ethyl lactate gradually decreased, while acetic, hexanoic, and lactic acid significantly increased (Huang et al., 2022). In another study, seven compounds were considered as potential aging markers of *Gujinggong* Baijiu (*Strong*-flavor) through headspace gas chromatography-ion mobility spectrometry combined with chemometrics (Chen et al., 2021). The concentration of ethyl hexanoate, propyl hexanoate, ethyl pentanoate and ethyl heptanoate were positively correlated with aging time, while ethyl acetate, 2-methyl-1-propanol and methylpropane were inverse.

The volatile profile of freshly distilled *Luzhoulaojiao* Baijiu (*Strong*-flavor) in two years aging was collected by headspace solid-phase microextraction coupled with GC–MS. Partial least-squares discriminant analysis, Spearman's correlation analysis and Random-forest decision-tree were selected to identify potential aging-markers. Fourteen compounds were regarded as aging markers and half of them were long chain fatty acid ethyl esters (LCFAEEs). LCFAEEs were undetectable at the beginning of aging because of their lower concentrations (Song et al., 2020). With the extension of aging time, LCFAEEs possessed longer carbon chains and higher concentrations. Ethyl oleate was proved to be the prominent single aging-marker. Baijiu is commonly documented to have colloidal properties (Zhang et al., 2021c). LCFAEEs with long alkane chains are prone to form stable colloidal clusters that could avoid the hydrolysis of corresponding esters.

The changes in aroma profile and key odorants of *Laowuzeng* Baijiu (*Laobaigan*-flavor) taken from the same batch and container during one-

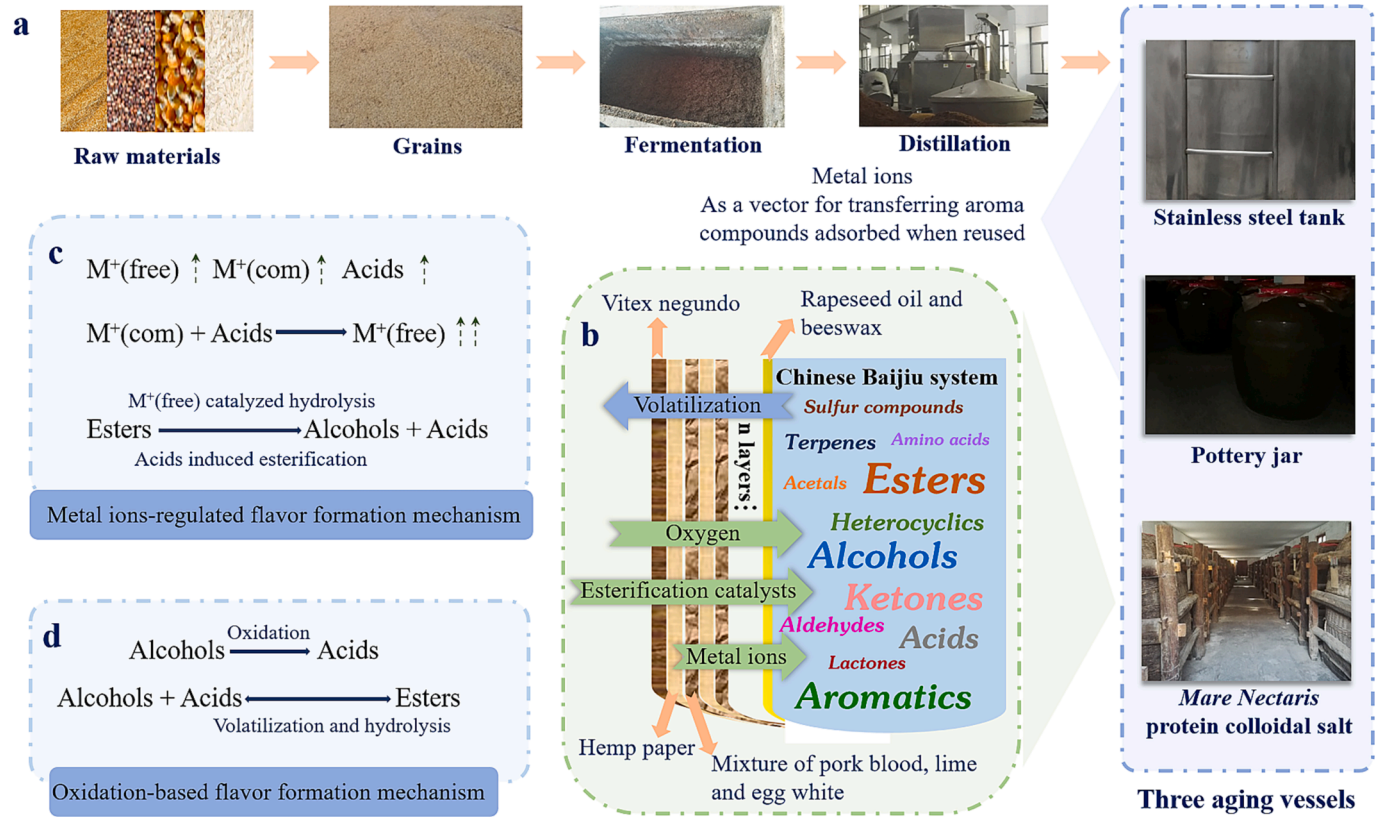


Fig. 2. Traditional production processing and aging mechanism of Baijiu. (a) Raw material is mixed uniformly with whole or powdered grains. The mixed grains are then fermented and distilled. The fresh Baijiu is then stored and aged in stainless steel vessels, *Mare Nectaris* or pottery jars. (b) Molecular mechanism of the role of *Mare Nectaris* in the aging process of Baijiu. (c) Metal ion-regulated flavor formation mechanism in the aging process of Baijiu. (d) The oxidation-based flavor formation mechanism in the aging process of Baijiu.

year aging period were explored. The satisfactory flavor characteristics, fruity, floral, acidic, sweet/honey and cheesy notes were enhanced during aging, while pickled vegetable, grain and alcoholic were impaired. The triangle test proved that "fruity" compounds, ethyl butanoate, ethyl hexanoate, ethyl octanoate, 3-methyl-1-butyl acetate, ethyl pentanoate, ethyl 3-methylbutanoate, ethyl acetate and ethyl lactate, were crucial to enhance the "aging" aroma (Zhu et al., 2020). 1,1-Diethoxymethane and methanethiol were identified as aging markers of *Jingzhi* Baijiu (*Sesame*-flavor). 1,1-Diethoxymethane is formed by acetalization of ethanol and formaldehyde and is only detected in aged Baijiu (Zhu, Fan, Xu, & Zhou, 2016). The concentration of 1,1-diethoxymethane is positively correlated with aging time, while methanethiol is on the contrary.

Natural and gamma irradiation aging process of *Feng*-flavor Baijiu were investigated by ultrahigh performance liquid chromatography Orbitrap mass spectrometry. Combined with foodomics, twenty-nine compounds were found to be related with aging process, concretely contain fifteen organic acids, eight esters and 6 other compounds (Jia et al., 2020c). Both the esters and organic acids showed an increasing trend in the aging process of *Feng*-flavor Baijiu. Esters are the most crucial aromatic compounds while acids are the fundamental flavor components of Baijiu (Xu et al., 2022). Besides, sixty compounds including organic acids and other flavor substances increased dramatically, fifteen unpleasant and harmful substances reduced after gamma irradiation.

Forty-eight *Feng*-flavor Baijiu samples of various aging years taken out from storage containers without blending were analyzed. The information of nonvolatile substances could be collected through ultrahigh performance liquid chromatography quadrupole-orbitrap high resolution mass spectrometry. Notably, non-volatiles were proved to be the precursors of volatiles. In total of 45 characteristic compounds were discovered through the application of statistical analysis, among them, 5-fluoro-2,3,4-three chlorobenzoic acid decreased, while the remaining compounds showed remarkable up-regulation as the aging years increased, specifically including ten esters, six organic acids, eight amino acids, eleven bioactive compounds and nine other components. The six non-volatile acids, guanosine butyric acid, creatine, cinnamic acid, malic acid, nicotic acid, and dihydroxy stearic acid, could promote Baijiu to be mellow and soft by utilizing recombination test (Jia et al., 2021a). Amino acids have a vital position for the flavor formation of Baijiu. Pyrazine with characteristic flavor is primarily produced by the degradation of nitrogen sources which contains amino acids through strecker reaction (Jia, Guo, Zhang, & Shi, 2023b).

The changes of specific components and aging markers of different flavor types of Baijiu during aging are different, but the overall trend is to endow Baijiu more harmonious flavor. The difference of different flavor Baijiu may be due to the difference of liquor samples themselves. The difference of the same flavor Baijiu may be due to the different storage containers or storage conditions that have different effects on the physical and chemical reactions in the liquor body, leading to the inconsistency of the dominant chemical reactions. With regard to the question of which substances can be used as aging markers of certain flavor Baijiu, it is necessary to conduct a lot of research on this flavor Baijiu to find the rules (Jia, Di, & Shi, 2023a).

3. Molecular mechanisms of Baijiu aging

The aging of Baijiu is a complicated and dynamic process, and is the result of various physical and chemical reactions. The aging mechanisms generally cover volatilization of irritant components with low boiling point; hydrogen bond association between alcohol and water; oxidation, esterification and hydrolysis among alcohols, aldehydes, acids and esters (Ma, Yi, & Zhu, 2019; Zheng et al., 2021a; Deng et al., 2020). In this part, more scientific molecular mechanisms of different traditional aging methods and artificial aging technologies will be clarified clearly based on the present research to promote comprehensive understanding of Baijiu aging process and development of novel artificial aging technology of Baijiu.

3.1. Traditional aging methods

Mare Nectaris, a unique aging container utilized in *Feng*-flavor Baijiu manufacture, is made of various raw materials containing wicker, hemp paper, pork blood, lime, egg white, rapeseed oil and beeswax. The container is a kind of protein colloidal salt which could efficiently bring about respiration to ensure beneficial component exchange between Baijiu system and external environment (Pang et al., 2018). The crucial role of *Mare Nectaris* in the aging process of Baijiu has been fully investigated (Fig. 2b). Metal ions and natural esterification catalysts contained in the raw materials of *Mare Nectaris* could migrate to Baijiu thus accelerate the esterification process. The characteristic amino acids and functional trace substances existed in aged *Feng*-flavor Baijiu were the consequence of dissolution and transformation of compounds in *Mare Nectaris* (Jia et al., 2021a). Moreover, compounds with –OH, –CHO, -C═O, –COOH, or $–NH_2$ groups could form colloid with centric ions in the modality of ligands which boost binding extent between ethanol and water molecules and facilitate Baijiu to a stable status.

Pottery vats and stainless vessels are the commonly adopted aging containers. Pottery vats are made of clay thus contain iron and copper ions, while Baijiu is stored in such containers, metal ions could permeate into Baijiu system during the aging period (Liu & Sun, 2018). Besides, almost all the distillation, cooler and delivery devices contain metal ions which could be corroded by organic acids exist in Baijiu during production (Huang et al., 2020). Metal ion-regulated flavor formation mechanism of aged Baijiu was clarified via multi-method analysis which consist of electrochemistry method and inductively coupled plasma mass spectrometry. Cyclic and square wave voltammetry confirmed that Baijiu system include both combination and free state of metal ions. During the aging period, the concentration of total metal ions and organic acids showed an upward trend and significant organometallic interactions performed between them (Zheng et al., 2021a). The increase of organic acids facilitated the transformation of metal ions from complex to free state (Fig. 2c). The free metal ions existed in Baijiu could catalyze the hydrolysis of esters, that means the esterification is a reversible reaction.

Esterification is the main reaction occurred during Baijiu aging process. A Pearson correlation analysis among three pair's compounds in *Laobaigan*-flavor Baijiu, acetic acid and ethyl acetate, hexanoic acid and ethyl hexanoate, 2-phenyl-1-ethanol and 2-phenylethyl acetate, was performed based on their concentrations during the aging process. The analyses clearly revealed that the concentrations of acetic acid, hexanoic acid and 2-phenyl-1-ethanol were highly positively correlated with the corresponding esters. This indicated that the esterification reaction is one of the complex aging mechanisms (Zhu et al., 2020). Flavor compounds of various aged Baijiu were analyzed by gas chromatography, based on the result, the component transformation among alcohols, acids, esters and relationships with vintage Baijiu were clearly elucidated by utilizing constituent analysis. It was worth noting that oxidation could increase the esters concentrations during aging by converting alcohols into corresponding acids and esters (Deng et al., 2020). The esterification reaction is reversible: if the esters concentration in fresh Baijiu is heavy, esters will volatilize and hydrolyze thus its concentration will be reduced; if the esters concentration is low, oxidation will facilitate the formation of esters so that the esterification reaction can reach balance.

The oxidation-based process holds a crucial position on the flavor formation of aged Baijiu. Natural aging includes a diversity of compound transformation, but the oxygen-based process is performed similar to electrochemical oxidation (Xiong et al., 2020). Three-electrode electrochemical system combined with ultrahigh performance liquid chromatography quadrupole-orbitrap high resolution mass spectrometry was used to investigate the oxidation-based flavor

formation mechanism. Electrochemical oxidation accelerated the transformation from alcohols to acids, thus increased the concentration of related esters that contribute to the flavor of vintage Baijiu (Fig. 2d). The oxidation of alcohols was the rate control step and electrochemical oxidation facilitated the oxidation of alcohols thus improved the concentrations of corresponding esters (Zheng et al., 2021b). Besides, gold electrode acted as a catalyst during the electrochemical oxidation. The unpaired d-orbital of gold electron ensured its catalytic activity.

The esterification and hydrolysis reaction mechanisms were also expounded at atomic- and molecular-level by density functional theory and molecular dynamics simulation. The activation energy values of alkyl-oxygen protonation for esters were lower than those of hydroxyl-oxygen protonation for corresponding acids which was consistent with the phenomenon that the concentrations of esters reduced but acids increased. The ethanol–water-acids system with stronger electrostatic energies and more hydrogen bonds was more prone to generate associative structures with flavor compounds during the aging period compared with the ethanol–water-esters system. The decrease of organoleptic stimulation of Baijiu is related to the hydrogen bond of components in the system (Huang et al., 2022). This indicated that acids are inclined to generate and strengthen associative structures during storage.

3.2. Artificial aging technologies

The strategy of soaking pottery chips or pottery powder in Baijiu was developed as an alternative aging technique. Trace components of Baijiu absorbed onto the surface of pottery powder could be the mechanism of aging but not the only. The particle size of pottery chips is an important factor affecting the sorption process, the adsorption capacity sharply decreases with the increase of particle size. The pseudo-second-order kinetic model could well describe the sorption kinetics of pottery powder for Baijiu micro-compounds and the external diffusion is the rate-limiting step. The flavor compounds of Baijiu including alcohols, esters, acids, and furan compounds existed in pottery powder in equilibrium state were characterized by GC–MS (Li, Fan, & Xu, 2021b). By and large, pottery powder not only affect the final sensory properties of Baijiu, but it can also as a vector to transferring flavor compounds adsorbed previously when reused.

Given the shortcoming of low productivity of conventional Baijiu aging process, there has been a trend towards physical technologies which could markedly shorten aging time and raise efficiency. Gamma irradiation had been utilized as an artificial aging technology. Irradiation mainly act on water in Baijiu to generate massive free radicals or molecular with high activity thus accelerate oxidation–reduction and esterification reactions occurred during aging. Irradiation treatment could reach the same state as natural aging and the aging time could be significantly shortened (Jia et al., 2020c). Besides, only the absorbed dose of gamma irradiation could influence the aging of Baijiu.

Magnetic field is another artificial aging strategy. Previous researches have resulted that non-uniform magnetic field condition was preferable to the aging of Baijiu than stable uniform magnetic field treatment. The influence of inhomogeneous alternating magnetic field on the aging of *Feng*-flavor Baijiu was lucubrated via untargeted foodomics and mass spectrometry analysis. The aging degree of magnetized samples increased with the elevation of magnetic field intensity and the maximum consequence was equal to 12.81 years of natural aging (Jia, Fan, Du, & Shi, 2021b). The adjustment of magnetic field intensity on Baijiu primarily exhibited by regulating the steric structure of molecules, adjusting the rate of Larmor precession, shortening the time of free radical transition from single to three lines state, lessening the solvent cage reaction rate of triplet radical pairs and strengthening the association of hydrogen bonds.

Ultra-high-pressure homogenization had been used as an artificial aging technology and its molecular mechanism was thoroughly expounded by utilizing foodomics combined with mass spectrum information optimization program based on colloid theory. Under the environment of high pressure, temperature and flow rate, the molecular arrangement occurred in the *Feng*-flavor Baijiu system because of mechanical shear and liquid layer friction. Concurrently, macromolecules were transformed into nanoscale colloidal particles thus boosted the formation of stable associative compounds in soliquid state and strengthened the mutual reaction among molecules. Besides, low-boiling compounds volatized simultaneously (Jia, Fan, Du, & Shi, 2022a). Eventually, the sensory characteristics and quality of Baijiu were enhanced in a shorter amount of time. The polynomial regression model verified that fresh distilled Baijiu could reach a natural aging state of 6.43 years under the condition of 400 bar.

4. Discrimination of Baijiu vintage

For the last few years, various brands, grades and vintage Baijiu were widely distributed in the market. In general, the common counterfeit method is the substitution of a shorter aging years Baijiu for another aging properly or adulteration with water or other inexpensive additives (Lin, Kang, Jin, Man, & Chen, 2020). Considering the above situation, the discrimination of Baijiu with different aging years is a significant portion of the protection of product quality and market supervision. Therefore, more facile, economical and quick methods for discriminating Baijiu are urgently needed.

The authentication and discrimination of Baijiu aging status has been performed by the combination of component characterization and multivariate analysis, then build a discrimination model. A gas-chromatography-flash electronic nose technique combined with chemometric analysis was developed for the identification of Chinese *Kaoliang* Baijiu of different aging times. The established predication model had an accuracy of 90 %. However, this model had lower accuracy for different brands of Chinese *Kaoliang* Baijiu, this is due to the different raw materials and manufacturing processes (Peng et al., 2017). The vintage authenticity of different flavor Baijiu, referring to the actual storage years from bottled, packaged to consumed in the market, was analyzed by gas chromatography (GC) and hydrogen proton nuclear magnetic resonance (^{1}H NMR) combined with multivariate statistical analysis. Partial least squares regression models were optimized and established to identify actual storage vintage of Baijiu based on various data processing methods and three variable selection methods which include Jack-knife P values, Jack-knife absolute coefficients and variable importance in projection (VIP) values. The components both detected in GC and ^{1}H NMR technologies were considered as potential aging markers, including acetaldehyde, butyrate, valeric acid, *n*-butanol, 2-butanol, hexanol, ethyl butyrate, ethyl valerate, ethyl heptanoate and ethyl caproate. Nevertheless, the concentration of selected aging markers is not linearly related to the aging years of Baijiu (Li et al., 2021c). The prediction model possessed excellent accuracy and can be regarded as an effective tool for vintage identification of Baijiu.

Headspace-gas chromatography-ion mobility spectrometry (HS-GC-IMS) combined with chemometrics were put forward as a strategy to identify the age of Baijiu. Thirty-nine *Strong*-flavor Baijiu samples with various vintage stored in pottery jars without blending were analyzed via HS-GC-IMS. Based on the acquired database, two discriminant models were established and verified for Baijiu aging time identification according to two hundred and twelve signal peaks (model A) and ninety-three identified compounds (model B) respectively, through partial least squares regression analysis. Herein, model B was more accurate to distinguish age-unknown Baijiu samples, exhibiting the goodness of fit value (R^2) of 0.9986 and root mean square error of prediction (RMSEP) of 0.244 (Chen et al., 2021). Besides, a prediction model on account of forty-five characteristic molecules of aged *Feng*-flavor Baijiu which were analyzed through ultrahigh performance liquid chromatography mass spectrometry combined with various statistical analysis and artificial neural networks was established to discriminate Baijiu vintage and the prediction accuracy was up to 100 % (Jia et al., 2021a). On the strength

of untargeted foodomics and mass spectrum information optimization program, a novel and accurate predication model for *Feng*-flavor Baijiu was furnished based on a polynomial regression model with R more than 0.9 and factor analysis with Kaiser-Meyer-Olkin value of 0.784 (Jia et al., 2022a). There existed a preferable relationship between potential distinct characteristic compounds and aging period.

Electrochemical methods have been a powerful tool to discriminate aged Baijiu with the advantage of no other sample preparation technique (Xiong et al., 2020). Moreover, electrochemical technologies could be more compatible to the analysis of Baijiu that without the addition of any redox-active agents. Baijiu with different aging times were identified through a triple-electrode system, ultraviolet–visible spectrometry and principal component analysis (PCA). The correlation between the reduction current and the total concentration of redox-active species was established by utilizing a polycrystalline gold-electrode with radius of 5 μm because of the mass transport rate increases along with the decrease of electrode size (Zhao et al., 2019). The intermolecular reactions between Baijiu components and ferrocenylmethanol ($FcCH_2OH$), an electrochemical label, was used to identify aged Baijiu through cyclic voltammetry. Because of the hydrogen bonding existing between $FcCH_2OH$ and ethanol (C_2H_5OH), the anodic and cathodic peak of $FcCH_2OH$ were both positively shifted (Hu et al., 2020). While only the anodic peak potential of $FcCH_2OH$ was positively shifted due to the formation of ion-pair between the oxidized form of $FcCH_2OH$ and the negatively charged carboxylate ($RCOO^-$). Besides, a visibly positive correlation was found between peak potential and the concentration of carboxylic acids present in Baijiu.

Baijiu is not a homogeneous solution but exist in a soliquid state that composed of abundant microcosmic multi-molecules (Wang et al., 2020). An effortless and non-disruptive approach for vintage Baijiu discrimination was demonstrated with electrochemical impedance spectrums (EIS) according to the distinctive colloidal impedance phenomena of Baijiu. EIS spectra then were simulated with the relevant equivalent circuit to receive parameter values which represent the physicochemical properties of aged Baijiu (Fig. 3). The resultant multi-elements data were dimensionally reduced by means of PCA. The PCA score plots showed that aged Baijiu can be separated commendably with a defined resolution. Besides, faradic impedance spectroscopy taking with graphite from the University of Idaho Thermolyzed Asphalt Reaction (GUITAR) electrode provides higher resolution than glassy carbon electrode EIS measurements (Jiang, Xie, Wan, Chen, & Zheng, 2019). The discriminating model mentioned above was built based on the differences in physiochemical properties of aged Baijiu other than its subtle flavor compounds variation, which ensured the reliability of the above methodology.

On account of the metal ion regulated deposition on gold nanorods (Au NP), a Baijiu colorimetric sensor array was proposed. Metal ions (Mg^{2+}, Pb^{2+}, Fe^{2+}, and Fe^{3+}) could adjust the microenvironment of Baijiu colloid to regulate the aggregation degree of Au NPs in the sensor array (Li et al., 2019). Besides, a colorimetric sensor array based on silver deposition on gold nanorods (AuNRs) was developed for the discrimination of different flavor-types Baijiu. Silver ions were reduced to Ag nanoshells on the surface of AuNRs by different reductants. AuNRs exhibit peculiar optical advantage and perform longitudinal localized surface plasmon resonance within the visible and near-infrared regions which endowing them extremely sensitive to tiny changes in the aspect ratio, so four different morphologies AuNRs were adopted. The silver deposition process can be inhibited by various compounds with hydroxyl or amino groups existed in Baijiu, thus providing different color fingerprints (Fig. 4a). The colorimetric sensor array showed the beneficial of accessible, fast-response and excellent discrimination. Moreover, it could be extended by adopting various size of AuNRs and different types of reductants (Jia et al., 2020a). These methods could be alternative tools for the identification of vintage Baijiu.

A novel four channel colorimetric sensor array based on the classic redox reaction between silver nitrate and *o*-phenylenediamine (OPD) or its three derivatives (OPDs) was constructed to determine the variety and concentrations of carbonyl flavor compounds existed in Baijiu. The detection mechanism is based on the fact that silver ions (Ag^+) and other oxidants could oxidize OPD or OPDs to different colored products (Wu et al., 2022). Different storage years of Baijiu possess different levels of carbonyl flavor compounds, thus can be successfully identified (Fig. 4b). The method was proved to be simple, rapid, reliable, and has good application potential (Zhang et al., 2021a). Six types of Baijiu with different cellar time of *Luzhoulaojiao* brand were successfully identified. Based on the previous researches, colorimetric sensor is another technique for the discrimination of Baijiu with simple, rapid and sensitive characteristics.

5. Challenges and perspectives

Owing to the diversity of raw materials, microorganisms, fermentation and aging processes during Baijiu production (Wei, Zou, Shen, & Yang, 2020), multitudinous flavor types of Baijiu had been defined and produced but the current research on aging process of Baijiu mainly focused on *Strong*-, *Light*- and *Feng*-flavor Baijiu (Table 1). Besides,

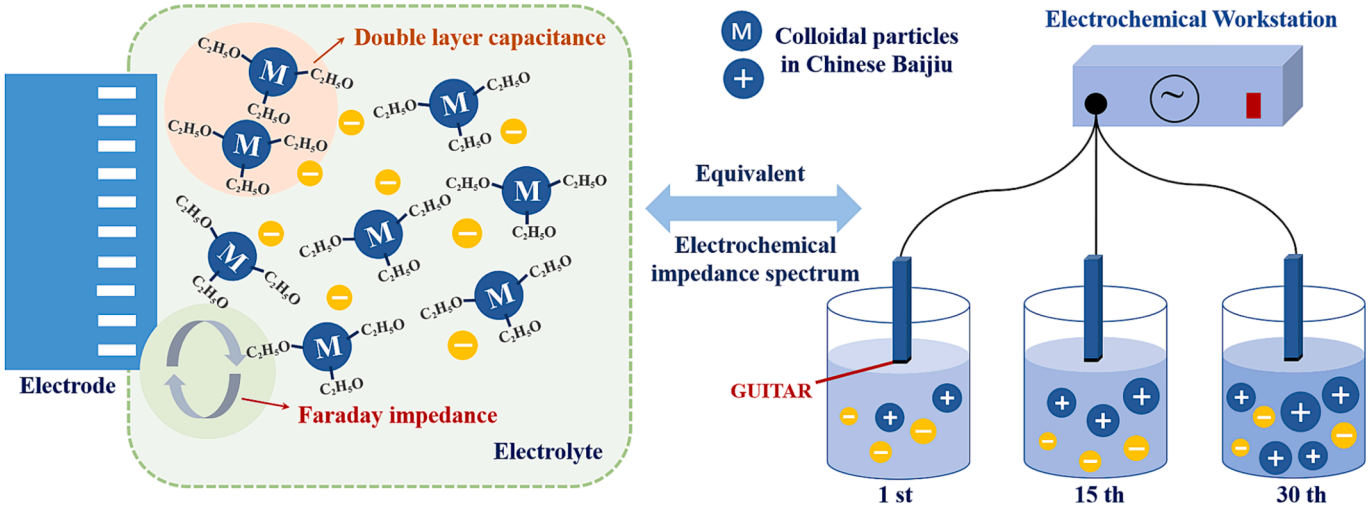


Fig. 3. The method to evaluate aging degree of Baijiu by measuring the electrochemical impedance spectrum (EIS) of verities' aged Baijiu according to the distinctive colloidal impedance phenomena of Baijiu. EIS spectra were simulated with the corresponding equivalent circuit (Jiang et al., 2019). GUITAR: graphite from the University of Idaho Thermolyzed Asphalt Reaction.

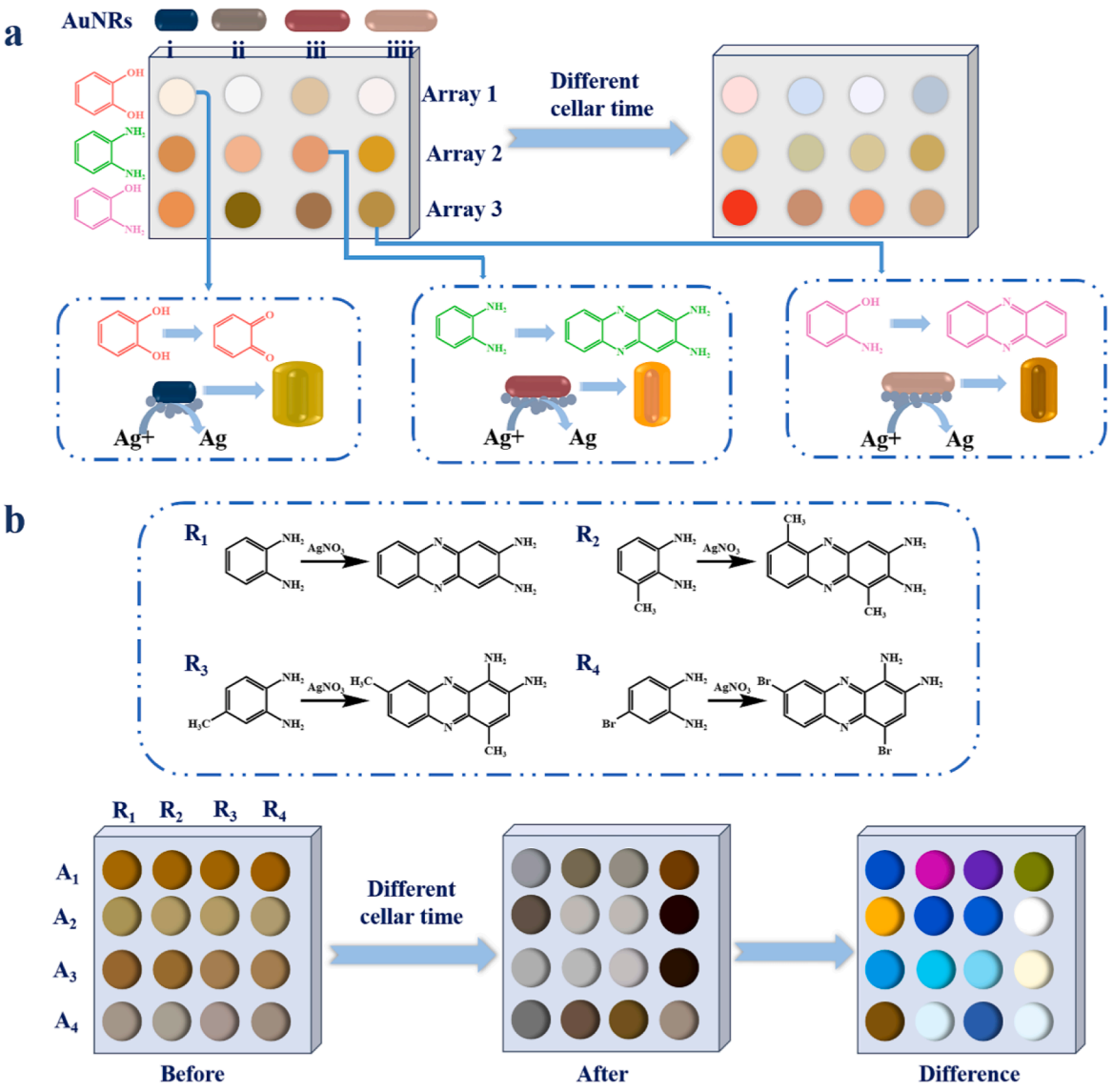


Fig. 4. Two different colorimetric sensor arrays for the discrimination of vintage Baijiu. (a) Illustration of colorimetric sensor array for the discrimination of Baijiu based on silver deposition of gold nanorods (AuNRs). i, ii, iii, iiii represent different sizes of AuNRs (Jia et al., 2020a). (b) Illustration of colorimetric sensor array for the discrimination of Baijiu based on silver nitrate and *o*-phenylenediamine derivatives. R_1, R_2, R_3, R_4 refer to different *o*-phenylenediamine derivatives (Wu et al., 2022).

certain non-volatile compounds are proved to be the precursors of volatile compounds or interact with other compounds thus affect the flavor of Baijiu (Hong, Tian, & Zhao, 2020). The present research mainly focused on volatile compounds' variation during aging period, the characterization of non-volatiles is lacked (Huang et al., 2019). Thus, a considerable amount of research on the Baijiu system variation during aging period need to be further investigated, especially various types of Baijiu to search for common rules and aging markers which could contribute to the quality control and improvement of Baijiu (Jia, Wang, & Shi, 2023c). In addition, the future research of Baijiu should be adhered to the dual guidance of flavor and health (Li, Yuan, Yong, Zhao, & Liu, 2020b).

The aging process is extremely complicated that involves various physical and chemical reactions (del Fresno et al., 2018; Tchabo et al., 2017). The mechanism of aging process cannot be fully explained by one or several theories, and the interaction between different mechanisms is more worthy of study. Traditional aging methods are time-consuming and labor-intensive which heavily affect the productivity and economic efficiency of Baijiu industry, utilizing artificial aging techniques has become an inevitable tendency (Solar, Castro, & Guerrero, 2021). The artificial aging technologies applied in wine were extensively researched including micro-oxygenation, oak products and external energy aging methods (Ma et al., 2022). The development of wine aging techniques could provide a specific direction and capital idea for the Baijiu aging procedure.

Micro-oxygenation refers to the simulation of microenvironment of oak barrel aging (Curko et al., 2021; Granja-Soares et al., 2020). Oak products are designed to simulate the oak extraction function of oak barrel aging process (Barbosa et al., 2022; Martinez-Gil, del Alamo-Sanza, Nevares, Sanchez-Gomez, & Gallego, 2020). External energy aging methods including ultrahigh pressure, pulsed electric fields, ultrasound fields, microwave fields and irradiation treatments. All of them

Table 1
Characterization methods and aging markers of different flavor types Baijiu.

Flavor type	Aging Baijiu samples	Characterization methods	Aging markers	Reference
Light-flavor	aged *Highland Qingke* Baijiu of 0–11 months	comprehensive two-dimensional gas chromatography-time-of-flight mass spectrometry	1,1-diethoxy propane, 1-(1-ethoxyethoxy)-propane, 1,1-diethoxy-butane, 1-(1-ethoxyethoxy)-butane, 1,1-diethoxy-3-methyl-butane, 1-(1-ethoxyethoxy)-hexane, 1-(1-ethoxyethoxy)-pentane, ethyl 3-methylbutyl ester, acetophenone, dihydro-4-hydroxy-2(3H)-furanone, 1,3,5-trioxane	Wang et al., 2022c
Light-flavor	0-year-old and 25-year-old *Xiaoqu* Baijiu	gas chromatography–olfactometry, gas chromatography-mass spectrometry, gas chromatography-flame ionization detector, gas chromatography-pulsed flame photometric detector	3-hydroxy-4,5-dimethyl-2(5H)-furanone (sotolon), vanillin, 3-(methylthio)-propionaldehyde (methional)	Sun et al., 2022
Light-flavor	aged *Niulanshan* base Baijiu of 3–13 years	gas chromatography–olfactometry-mass spectrometry, gas chromatography-mass spectrometry, gas chromatography-flame ionization detector	ethyl acetate, ethyl acrylate, ethyl 2-methylbutyrate, γ-nonalactone, ethyl isovalerate, ethyl butyrate, isoamyl acetate, ethyl caprylate, ethyl valerate, 3-methylbutanal, β-damascenone, geosmin	Wang et al., 2022d
Strong-flavor	aged Baijiu from 1998 to 2019	headspace-gas chromatography-ion mobility spectrometry	ethyl hexanoate, propyl hexanoate, ethyl pentanoate, ethyl heptanoate, ethyl acetate, 2-methyl-1-propanol, and methylpropane	Chen et al., 2021
Strong-flavor	aged *Lu*-flavor Baijiu of 0–4 years	headspace solid-phase micro-extraction coupled with gas chromatography-mass spectrometry	ethyl oleate, 2-phenylethyl hexanoate, hexanoic acid, ethyl linoleate, isobutyric acid, phenethyl ester, ethyl hexadec-9-enoate, 2,6-di-*tert*-butyl-4-methylphenol, 1-undecanol, ethyl laurate, ethyl pentadecanoate, ethyl palmitate, ethyl myristate, 2,4-di-*tert*-butylphenol, isobutyraldehyde	Zhang et al., 2021c
Sesame-flavor	aged raw Baijiu from 1985 to 2014	headspace solid-phase micro extraction coupled with gas chromatography-mass spectrometry	1,1-diethoxymethane, methanethiol	Zhu et al., 2016

are based on the mechanism that supplied energy could be transformed into the activation energy of reaction needed in the aging process (Gao et al., 2021; Christofi, Malliaris, Katsaros, Panagou, & Kallithraka, 2020; Arcena et al., 2022). Nevertheless, unclear reaction molecular mechanisms and principles put the artificial aging technologies into a theoretical research stage and restrict the commercialization of artificial aging methods. It is imperative to take more efforts to expound the underlying aging mechanisms thus regulate and optimize the processing conditions.

The most common strategy for discriminating vintage Baijiu is based on the difference of composition. The conventional chromatography and mass spectrometry characterizations are precise but accompanied with inevitable shortcomings such as complicated operation, time-consuming and high cost (Jia et al., 2020b). Electrochemical method and colorimetric sensor array have been powerful tools to discriminate aged Baijiu with the advantage of simple, rapid, sensitive and no other sample pretreatment (Dai et al., 2021; Li et al., 2022). The in-depth research of unique colloidal impedance behavior of Baijiu could be a future direction of development due to its powerful effect on the understanding of aging mechanisms. In order to better supervise Baijiu market, the situ real-time measurements are urgently needed which could promote the development of more easy-operation, time-saving and low-cost identification strategies.

6. Conclusions

The variation of Baijiu system during aging period, aging mechanisms and discrimination strategies for Baijiu with various vintages were systematically illustrated in the review. The detailed composition changes and aging markers of different flavor-types of Baijiu are different during aging process, yet the general tendency is consistent. The aging mechanisms of Baijiu can be described as the synergy of physicochemical reactions including volatilization, association, hydrolysis, oxidation and esterification catalyzed by metal ions, other raw materials dissolved from storage vessels or compounds in Baijiu. The authentication and discrimination of Baijiu vintage has been performed by component characterization coupled with multivariate analysis, electrochemical method and colorimetric sensor array. Nevertheless, more in-depth research of the above three aspects is imperative which could lay the foundation for the improvement of Baijiu artificial aging technologies and Baijiu quality.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments

This work was financially supported by National Key Research and Development Program of China (No. 2019YFC1606300), Key R&D technology projects in Shaanxi Province (No. 2021NY-185), Xianyang Science and Technology Plan Project (No. 2021ZDYF-NY-0025), and Xi'an Science and Technology Plan Project (No. 21NYYF0056).

References

Arcena, M. R., Leong, S. Y., Then, S., Hochberg, M., Sack, M., Mueller, G., … Oey, I. (2022). The effect of pulsed electric fields pre-treatment on the volatile and phenolic profiles of Merlot grape musts at different winemaking stages and the sensory characteristics of the finished wines. *Innovative Food Science & Emerging Technologies, 70*, Article 102698. https://doi.org/10.1016/j.ifset.2021.102698

Barbosa, R. B., Santiago, W. D., Alvarenga, G. F., Oliveira, R. E. D., Ferreira, V. R. F., Nelson, D. L., & Cardoso, M. D. (2022). Physical-chemical profile and quantification of phenolic compounds and polycyclic aromatic hydrocarbons in cachaça samples aged in oak *(Quercus* sp.) barrels with different heat treatments. *Food and Bioprocess Technology, 15*, 1977–1987. https://doi.org/10.1007/s11947-022-02853-w

Chen, S., Lu, J. L., Qian, M. C., He, H. K., Li, A. J., Zhang, J., … Xu, Y. (2021). Untargeted headspace-gas chromatography-ion mobility spectrometry in combination with chemometrics for detecting the age of Chinese liquor (Baijiu). *Foods, 10*, 2888. https://doi.org/10.3390/foods10112888

Christofi, S., Malliaris, D., Katsaros, G., Panagou, E., & Kallithraka, S. (2020). Limit SO_2 content of wines by applying High Hydrostatic Pressure. *Innovative Food Science & Emerging Technologies, 62*, Article 102342. https://doi.org/10.1016/j.ifset.2020.102342

Curko, N., Ganic, K. K., Tomasevic, M., Gracin, L., Jourdes, M., & Teissedre, P. L. (2021). Effect of enological treatments on phenolic and sensory characteristics of red wine during aging: Micro-oxygenation, sulfur dioxide, iron with copper and gelatin fining. *Food Chemistry, 339*, Article 127848. https://doi.org/10.1016/j.foodchem.2020.127848

Dai, H. P., Jia, J. J., Fan, Y., Chen, H. Y., Wang, S. T., Shen, C. H., … She, Y. B. (2021). Four-channel fluorescent sensor array based on various functionalized CdTe quantum dots for the discrimination of Chinese baijiu. *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy, 252*, Article 119513. https://doi.org/10.1016/j.saa.2021.119513

del Fresno, J. M., Loira, I., Morata, A., Gonzalez, C., Suarez-Lepe, J. A., & Cuerda, R. (2018). Application of ultrasound to improve lees ageing processes in red wines. *Food Chemistry, 261*, 157–163. https://doi.org/10.1016/j.foodchem.2018.04.041

Deng, Y. H., Xiong, A., Zhao, K., Hu, Y. R., Kuang, B. S., Xiong, X., … Zheng, Q. (2020). Mechanisms of the regulation of ester balance between oxidation and esterification in aged Baijiu. *Scientific Reports, 10*, 17169. https://doi.org/10.1038/s41598-020-74423-z

Dong, W., Guo, R. N., Liu, M., Shen, C. H., Sun, X. T., Zhao, M. M., … Wu, J. H. (2019). Characterization of key odorants causing the roasted and mud-like aromas in strong-aroma types of base Baijiu. *Food Research International, 125*, Article 108546. https://doi.org/10.1016/j.foodres.2019.108546

Gao, X. L., Feng, T., Liu, E. M., Shan, P., Zhang, Z. K., Liao, L., & Ma, H. L. (2021). Ougan juice debittering using ultrasound-aided enzymatic hydrolysis: Impacts on aroma and taste. *Food Chemistry, 345*, Article 128767. https://doi.org/10.1016/j.foodchem.2020.128767

He, Y. X., Chen, S., Tang, K., Qian, M., Yu, X. W., & Xu, Y. (2021). Sensory characterization of Baijiu pungency by combined time-intensity (TI) and temporal dominance of sensations (TDS). *Food Chemistry, 147*, Article 110493. https://doi.org/10.1016/j.foodres.2021.110493

Hong, J. X., Tian, W. J., & Zhao, D. R. (2020). Research progress of trace components in sesame-aroma type of baijiu. *Food Research International, 137*, Article 109695. https://doi.org/10.1016/j.foodres.2020.109695

Hong, J. X., Zhao, D. R., & Sun, B. G. (2021). Research progress on the profile of trace components in Baijiu. *Food Reviews International*. https://doi.org/10.1080/87559129.2021.1936001

Hu, Y. R., Zhao, K., Gao, F., Zhang, J., Xu, D. X., Yang, Z. L., … Zheng, Q. (2020). Modulated interactions between the electrochemical label and liquor compounds: Differentiating between aging times of typical Chinese liquors. *Journal of The Electrochemical Society, 167*, Article 027517. https://doi.org/10.1149/1945-7111/ab68cf

Huang, M. Q., Huo, J. Y., Wu, J. W., Zhao, M. M., Sun, J. Y., Zheng, F. P., … Li, H. H. (2019). Structural characterization of a tetrapeptide from Sesame flavor-type Baijiu and its interactions with aroma compounds. *Food Research International, 119*, 733–740. https://doi.org/10.1016/j.foodres.2018.10.055

Huang, Z. J., Zeng, Y. H., Liu, W. H., Wang, S. T., Shen, C. H., & Shi, B. (2020). Effects of metals released in strong-flavor baijiu on the evolution of aroma compounds during storage. *Food Science & Nutrition, 8*(4), 1904–1913. https://doi.org/10.1002/fsn3.1475

Huang, Z. J., Zeng, Y. H., Sun, Q. Y., Zhang, W. H., Wang, S. T., Shen, C. H., & Shi, B. (2022). Insights into the mechanism of flavor compound changes in strong flavor baijiu during storage by using the density functional theory and molecular dynamics simulation. *Food Chemistry, 373*, Article 131522. https://doi.org/10.1016/j.foodchem.2021.131522

Jia, J. J., Wu, M. X., Wang, S. T., Wang, X. C., Hu, Y., Chen, H. Y., … She, Y. B. (2020a). Colorimetric sensor array based on silver deposition of gold nanorods for discrimination of Chinese white spirits. *Sensors and Actuators B: Chemical, 320*, Article 128256. https://doi.org/10.1016/j.snb.2020.128256

Jia, W., Du, A., Fan, Z. B., Zhang, R., Li, Y. L., Shi, Q. Y., … Chu, X. G. (2021a). Molecular mechanism of the role of Mare Nectaris in the Feng-Flavor Baijiu aging. *LWT - Food Science and Technology, 135*, Article 110254. https://doi.org/10.1016/j.lwt.2020.110254

Jia, W., Di, C. N., & Shi, L. (2023a). Applications of lipidomics in goat meat products: Biomarkers, structure, nutrition interface and future perspectives. *Journal of Proteomics, 270*, Article 104753. https://doi.org/10.1016/j.jprot.2022.104753

Jia, W., Fan, Z. B., Du, A., Li, Y. L., Zhang, R., Shi, Q. Y., … Chu, X. G. (2020b). Recent advances in Baijiu analysis by chromatography based technology-A review. *Food Chemistry, 324*, Article 126899. https://doi.org/10.1016/j.foodchem.2020.126899

Jia, W., Fan, Z. B., Du, A., & Shi, L. (2022a). Molecular mechanism of high pressure shear grinding on Feng-flavour Chinese Baijiu ageing. *Food Research International, 153*, Article 110957. https://doi.org/10.1016/j.foodres.2022.110957

Jia, W., Fan, Z. B., Du, A., & Shi, L. (2021b). Untargeted foodomics reveals molecular mechanism of magnetic field effect on Feng-flavor Baijiu ageing. *Food Research International, 149*, Article 110681. https://doi.org/10.1016/j.foodres.2021.110681

Jia, W., Fan, Z. B., Du, A., Shi, L., & Ren, J. M. (2022b). Characterisation of key odorants causing honey aroma in Feng-flavour Baijiu during the 17-year ageing process by multivariate analysis combined with foodomics. *Food Chemistry, 374*, Article 131764. https://doi.org/10.1016/j.foodchem.2021.131764

Jia, W., Guo, A. A., Zhang, R., & Shi, L. (2023b). Mechanism of natural antioxidants regulating advanced glycosylation end products of Maillard reaction. *Food Chemistry, 404*, Article 134541. https://doi.org/10.1016/j.foodchem.2022.134541

Jia, W., Li, Y. L., Du, A., Fan, Z. B., Zhang, R., Shi, L., … Chu, X. G. (2020c). Foodomics analysis of natural aging and gamma irradiation maturation in Chinese distilled Baijiu by UPLC-Orbitrap-MS/MS. *Food Chemistry, 315*, Article 126308. https://doi.org/10.1016/j.foodchem.2020.126308

Jia, W., Wang, X., & Shi, L. (2023c). Endogenous hydrocortisone caused metabolic perturbation and nutritional deterioration of animal-derived food in a dose-dependent manner. *Food Chemistry, 401*, Article 134145. https://doi.org/10.1016/j.foodchem.2022.134145

Jiang, X. Y., Xie, Y. Q., Wan, D. J., Chen, M. B., & Zheng, F. P. (2019). GUITAR-enhanced facile discrimination of aged Chinese Baijiu using electrochemical impedance spectroscopy. *Analytica Chimica Acta, 1059*, 36–41. https://doi.org/10.1016/j.aca.2019.01.050

Li, A. L., Jia, J. J., Fan, Y., Chen, H. Y., Wang, S. T., Shen, C. H., … She, Y. B. (2021a). Furfural and organic acid targeted carbon dot sensor array for the accurate identification of Chinese baijiu. *Journal of Food Science, 86*(7), 2924–2938. https://doi.org/10.1111/1750-3841.15766

Li, J. W., Liu, X. F., Luo, H. B., Zhao, D., Zhang, J., Qiao, C. L., … Hou, C. J. (2022). A gold nanorods etching based colorimetric sensor array for the detection of reducing substances and discrimination of Chinese Baijiu. *Food Analytical Methods*. https://doi.org/10.1007/s12161-022-02269-4

Li, J. W., Shui, Z. F., Dong, L., Shen, L., Zhao, D., Luo, H. B., … Huo, D. Q. (2020a). A novel acid-sensitive quantum dot sensor array for the identification of Chinese baijiu. *Analytical Methods, 12*, 4789–4797. https://doi.org/10.1039/D0AY01454K

Li, J. W., Yang, P., Zhang, J., Shui, Z. F., Huo, D. Q., Hou, C. J., … Shen, C. H. (2019). A metal ion-regulated colorimetric sensor array: Discriminating Chinese Baijiu from other beverage. *Analytical Methods, 11*, 5274–5281. https://doi.org/10.1039/C9AY01804B

Li, M., Fan, W. L., & Xu, Y. (2021b). Volatile compounds sorption during the aging of Chinese Liquor (Baijiu) using Pottery Powder. *Food Chemistry, 345*, Article 128705. https://doi.org/10.1016/j.foodchem.2020.128705

Li, Y. A., Yuan, S. Q., Yong, X. H., Zhao, T., & Liu, J. (2020b). Research progress on small peptides in Chinese Baijiu. *Journal of Functional Foods, 72*, Article 104081. https://doi.org/10.1016/j.jff.2020.104081

Li, Y. C., Fan, S. X., Li, A. J., Liu, G. Y., Lu, W., Yang, B., … Zhong, Q. D. (2021c). Vintage analysis of Chinese Baijiu by GC and ^{1}H NMR combined with multivariable analysis. *Food Chemistry, 360*, Article 129937. https://doi.org/10.1016/j.foodchem.2021.129937

Lin, H., Kang, W. C., Jin, H. J., Man, Z. X., & Chen, Q. S. (2020). Discrimination of Chinese Baijiu grades based on colorimetric sensor arrays. *Food Science and Biotechnology, 29*, 1037–1043. https://doi.org/10.1007/s10068-020-00757-z

Liu, H. L., & Sun, B. G. (2018). Effect of fermentation processing on the flavor of Baijiu. *Journal of Agricultural and Food Chemistry, 66*(22), 5425–5432. http://orcid.org/0000-0002-7912-2611.

Ma, N., Yi, F., & Zhu, J. (2019). Characterization of aroma-active compounds and perceptual interaction between esters and sulfur compounds in Xi baijiu. *European Food Research and Technology, 246*, 2517–2535. https://doi.org/10.1007/s00217-020-03594-w

Ma, T. T., Wang, J. Q., Wang, H. L., Zhao, Q. Y., Zhang, F., Ge, Q., … Sun, X. Y. (2022). Wine aging and artificial simulated wine aging: Technologies, applications, challenges, and perspectives. *Food Research International, 153*, Article 110953. https://doi.org/10.1016/j.foodres.2022.110953

Martinez-Gil, A. M., del Alamo-Sanza, M., Nevares, I., Sanchez-Gomez, R., & Gallego, L. (2020). Effect of size, seasoning and toasting level of *Quercus pyrenaica* Willd. wood on wine phenolic composition during maturation process with micro-oxygenation. *Food Research International, 128*, Article 108703. https://doi.org/10.1016/j.foodres.2019.108703

Pang, X. N., Han, B. Z., Huang, X. N., Zhang, X., Hou, L. F., Cao, M., … Chen, J. Y. (2018). Effect of the environment microbiota on the flavour of light-flavour *Baijiu* during spontaneous fermentation. *Scientific Reports, 8*, 3396. https://doi.org/10.1038/s41598-018-21814-y

Peng, Q., Xu, X., Xing, W. H., Hu, B. W., Shen, C., Tian, R. G., … Xie, G. F. (2017). Ageing status characterization of Chinese spirit using scent characteristics combined with chemometric analysis. *Innovative Food Science & Emerging Technologies, 44*, 212–216. https://doi.org/10.1016/j.ifset.2017.04.006

Solar, S., Castro, R., & Guerrero, E. D. (2021). New accelerating techniques applied to the ageing of oenological products. *Food Reviews International*. https://doi.org/10.1080/87559129.2021.1934009

Song, X. B., Jing, S., Zhu, L., Ma, C. F., Song, T., Wu, J. H., … Chen, F. (2020). Untargeted and targeted metabolomics strategy for the classification of strong aroma-type baijiu (liquor) according to geographical origin using comprehensive two-dimensional gas chromatography-time-of-flight mass spectrometry. *Food Chemistry, 314*, Article 126098. https://doi.org/10.1016/j.foodchem.2019.126098

Sun, J. Y., Wang, Z., & Sun, B. G. (2021). Low quantity but critical contribution to flavor: Review of the current understanding of volatile sulfur-containing compounds in Baijiu. *Journal of Food Composition and Analysis, 103*, Article 104079. https://doi.org/10.1016/j.jfca.2021.104079

Sun, X. Z., Qian, Q. Q., Xiong, Y. Q., Xie, Q. Q., Yue, X. X., Liu, J. H., … Yang, Q. (2022). Characterization of the key aroma compounds in aged Chinese Xiaoqu Baijiu by means of the sensomics approach. *Food Chemistry, 384*, Article 132452. https://doi.org/10.1016/j.foodchem.2022.132452

Tan, Y. W., Zhong, H. P., Zhao, D., Du, H., & Xu, Y. (2019). Succession rate of microbial community causes flavor difference in strong-aroma Baijiu making process. *International Journal of Food Microbiology, 311*, Article 108350. https://doi.org/10.1016/j.ijfoodmicro.2019.108350

Tchabo, W., Ma, Y. K., Kwaw, E., Zhang, H. N., Xiao, L. L., & Tahir, H. E. (2017). Aroma profile and sensory characteristics of a sulfur dioxide-free mulberry (*Morus nigra*) wine subjected to non-thermal accelerating aging techniques. *Food Chemistry, 232*, 89–97. https://doi.org/10.1016/j.foodchem.2017.03.160

Tian, T. T., Ruan, S. L., Zhao, Y. P., Li, J. M., Yang, C., & Cao, H. (2022). Multi-objective evaluation of freshly distilled brandy: Characterisation and distribution patterns of key odour-active compounds. *Food Chemistry: X, 14*, Article 100276. https://doi.org/10.1016/j.fochx.2022.100276

Wang, G. N., Song, X. B., Zhu, L., Li, Q., Zheng, F. P., Geng, X. J., … Sun, B. G. (2022a). A flavoromics strategy for the differentiation of different types of Baijiu according to the non-volatile organic acids. *Food Chemistry, 374*, Article 131641. https://doi.org/10.1016/j.foodchem.2021.131641

Wang, J. S., Chen, H., Wu, Y. S., & Zhao, D. R. (2022b). Uncover the flavor code of strong-aroma baijiu: Research progress on the revelation of aroma compounds in strong-aroma baijiu by means of modern separation technology and molecular sensory evaluation. *Journal of Food Composition and Analysis, 109*, Article 104499. https://doi.org/10.1016/j.jfca.2022.104499

Wang, L. L., Fan, S. S., Yan, Y., Yang, L., Chen, S., & Xu, Y. (2020). Characterization of potent odorants causing a pickle-like off-odor in Moutai-aroma type Baijiu by comparative aroma extract dilution analysis, quantitative measurements, aroma Addition, and omission studies. *Journal of Agricultural and Food Chemistry, 68*(6), 1666–1677. https://doi.org/10.1021/acs.jafc.9b07238

Wang, X. L., Song, X. B., Zhu, L., Geng, X. J., Zheng, F. P., Zhao, Q. Z., … Sun, B. G. (2022c). Unraveling the acetals as ageing markers of Chinese Highland Qingke Baijiu using comprehensive two dimensional gas chromatography-time-of-flight mass spectrometry combined with metabolomics approach. *Food Quality and Safety, 5*, fyab014. https://doi.org/10.1093/fqsafe/fyab014

Wang, Z., Wang, Y., Zhu, T. T., Wang, J., Huang, M. Q., Wei, J. W., … Meng, N. (2022d). Characterization of the key odorants and their content variation in Niulanshan Baijiu with different storage years using flavor sensory omics analysis. *Food Chemistry, 376*, Article 131851. https://doi.org/10.1016/j.foodchem.2021.131851

Wei, Y., Zou, W., Shen, C. H., & Yang, J. G. (2020). Basic flavor types and component characteristics of Chinese traditional liquors: A review. *Journal of Food Science, 85* (12), 4096–4107. https://doi.org/10.1111/1750-3841.15536

Wu, M. X., Chen, H. Y., Fan, Y., Wang, S. T., Hu, Y., Liu, J., … She, Y. B. (2022). Carbonyl flavor compound-targeted colorimetric sensor array based on silver nitrate and o-phenylenediamine derivatives for the discrimination of Chinese Baijiu. *Food Chemistry, 372*, Article 131216. https://doi.org/10.1016/j.foodchem.2021.131216

Xiong, A. Y., Zhao, K., Hu, Y. R., Yang, G. P., Kuang, B. S., Xiong, X., … Zheng, Q. (2020). Influence of electrochemical oxidation on the maturation process of the distilled spirit. *ACS Omega, 5*(29), 18349–18355. https://doi.org/10.1021/acsomega.0c02090

Xu, Y. Q., Zhao, J. R., Liu, X., Zhang, C. S., Zhao, Z. G., Li, X. T., & Sun, B. G. (2022). Flavor mystery of Chinese traditional fermented baijiu: The great contribution of ester compounds. *Food Chemistry, 369*, Article 130920. https://doi.org/10.1016/j.foodchem.2021.130920

Zhang, J., Huang, R., Zhang, J. J., Zhou, J., Qin, H., Yang, P., … Huo, D. Q. (2021a). A novel colorimetric sensor array for Baijiu discrimination based on gold triangular nanoplates etching. *Food Chemistry, 337*, Article 129789. https://doi.org/10.1016/j.snb.2021.129789

Zhang, S., Pu, J., Wen, J., She, D., Jang, A., Yue, P., & Du, J. (2021b). Effects of different storage containers on flavor components of Baijiu. *Liquor-Making. Science & Technology, 07*, 65–69. https://doi.org/10.13746/j.njkj.2021092

Zhang, X. J., Meng, L. J., Lu, Z. M., Chai, L. J., Wang, S. T., Shi, J. S., … Xu, Z. H. (2021c). Identification of age-markers based on profiling of Baijiu volatiles over a two-year maturation period: Case study of Lu-flavor Baijiu. *LWT - Food Science and Technology, 141*, Article 110913. https://doi.org/10.1016/j.lwt.2021.110913

Zhao, K., Liu, L. L., Zheng, Q., Gao, F., Chen, X. P., Yang, Z. L., … Yu, Y. G. (2019). Differentiating between ageing times of typical Chinese liquors by steady-state microelectrode voltammetry. *Microchemical Journal, 151*, Article 104244. https://doi.org/10.1016/j.microc.2019.104244

Zheng, Q., Hu, Y. R., Xiong, A., Su, Y., Wang, Z. H., Zhao, K., & Yu, Y. G. (2021a). Elucidating metal ion-regulated flavour formation mechanism in the aging process of Chinese distilled spirits (Baijiu) by electrochemistry, ICP-MS/OES, and UPLC-Q-Orbitrap-MS/MS. *Food & Function, 12*, 8899–8906. https://doi.org/10.1039/D1FO01505B

Zheng, Q., Wang, Z. H., Xiong, A. Y., Hu, Y. R., Su, Y., Zhao, K., & Yu, Y. G. (2021b). Elucidating oxidation-based flavour formation mechanism in the aging process of Chinese distilled spirits by electrochemistry and UPLC-Q-Orbitrap-MS/MS. *Food Chemistry, 355*, Article 129596. https://doi.org/10.1016/j.foodchem.2021.129596

Zhu, L., Wang, X. L., Song, X. B., Zheng, F. P., Li, H. H., Chen, F., … Zhang, F. Y. (2020). Evolution of the key odorants and aroma profiles in traditional *Laowuzeng* baijiu during its one-year ageing. *Food Chemistry, 310*, Article 125898. https://doi.org/10.1016/j.foodchem.2019.125898

Zhu, M. X., Fan, W. L., Xu, Y., & Zhou, Q. Y. (2016). 1,1-Diethoxymethane and methanethiol as age markers in Chinese roasted-sesame-like aroma and flavour type liquor. *European Food Research and Technology, 242*, 1985–1992. https://doi.org/10.1007/s00217-016-2697-x